Chemistry of AlGaN Particulate Formation

by **M. E. Coltrin and J. R. Creighton**

Motivation—GaN and AlGaN alloys are extremely important materials with widespread applications for optoelectronics (e.g. solid state lighting) and high power electronics. The growth of AlGaN thin films by metalorganic chemical vapor phase deposition (MOCVD) is often complicated by parasitic gas-phase chemical reactions that diminish the deposition efficiency and make it difficult to control alloy composition. The growth rate and alloy composition depend sensitively on temperature and other reactor variables, making the process difficult to control and optimize. Our earlier work showed that gas-phase particulates form during AlGaN MOCVD, and are responsible for many of these growth complications.

Accomplishment—We have used a combination of experiments and reactor modeling to investigate parasitic chemical reactions that occur during AlGaN MOCVD. Growth rates for GaN, AlN, and AlGaN were measured over a wide range of reactor conditions. Our results indicate that the parasitic chemical reactions require high temperatures and occur in the boundary layer near the growing surface. These reactions ultimately lead to the formation of nanoparticles, which we have recently observed using *in situ* laser light scattering. Thermophoresis keeps the nanoparticles from reaching the surface, so the material tied up in nanoparticles cannot participate in the thin film deposition process. We have developed a relatively simple, 9-reaction mechanism in which activated (i.e., temperature-dependent) chemical reactions form reactive intermediate chemical species that go on to nucleate the gas-phase particles. Once the particles have been formed, we propose that the particles grow further by an MOCVD-type mechanism, which is similar to the mechanism for the (desired) thin-film growth. The chemistry model includes steps describing Ga-precursor decomposition, Al-adduct formation and methane elimination, particulate nucleation, and particle growth in the AlGaN system. Reacting flow simulations were used to predict film growth rates and were compared with our rotating-disk reactor experiments. As seen in Fig. 1, the measured AlGaN growth rate drops with increasing temperature (a signature of the activated parasitic chemistry) and with increasing Al-precursor flow rate. Our model calculates both trends quantitatively. The solid AlGaN composition as a function of growth temperature is shown in Fig. 2. The aluminum fraction of the alloy, $X_s(Al)$, increases with inlet gas mole fraction, $X_g(Al)$, but is always smaller in magnitude. As temperature is increased, and thus as the parasitic reaction pathways become more and more important, the ratio $X_s(Al)$ / $X_g(Al)$ gets significantly smaller. Our model reproduces the general trends exhibited in Fig. 2, although not the strength of the curvature at the highest Al/III ratio.

Significance—The non-linear and non-ideal behavior of the AlGaN MOCVD has greatly impeded the growth and utilization of AlGaN material. Our investigations of the chemical mechanisms responsible for parasitic particle growth have led to a quantitative computer model of the growth process. This model will enable further reactor design modifications and process optimization to improve AlGaN growth and control of material quality.

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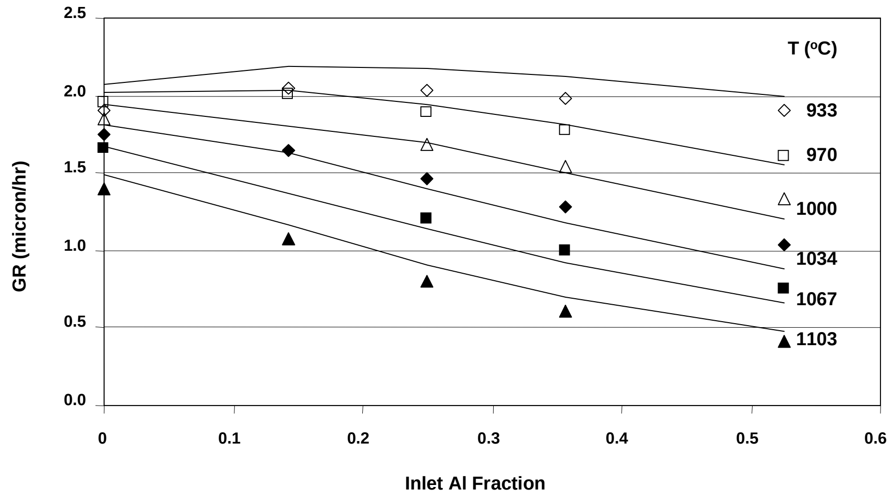

Figure 1. AlGaN growth rate as a function of growth temperature and Al/III (Al flow divided by total Group-III flow).

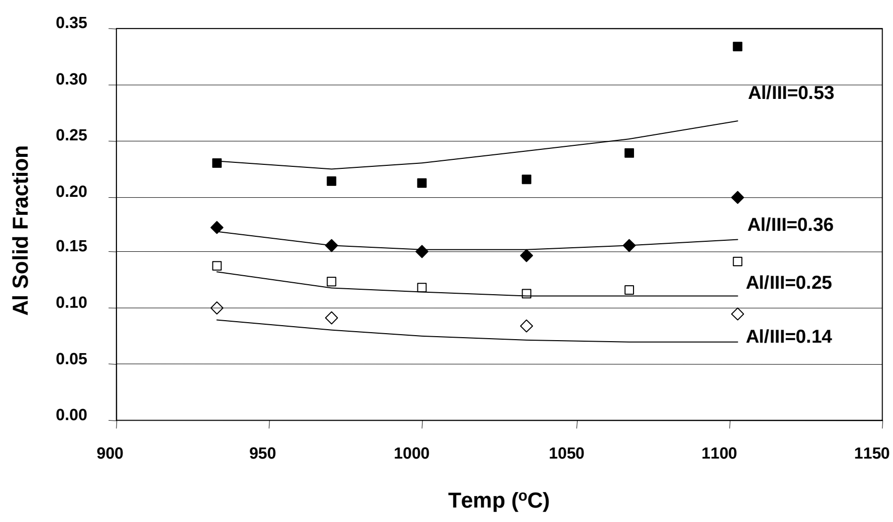

Figure 2. Al fraction in AlGaN alloy as a function of growth temperature and gas-phase Al fraction (Al flow divided by total Group-III flow).